Fundamental Studies of Solidification Using Real-Time X-Ray Microscopy

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Physical processes which occur at, or near, the solid-liquid interface during solidification or other phase transformations, partially determine important properties of solids. To date, interfacial morphologies and particle-interface interactions in the respective metallic, optically opaque systems have been deduced from post-process metallographic analyses of specimens. Thus, little information is obtained about the detailed dynamics of the processes.

We are developing a high-resolution x-ray microscope to view, in-situ and in real-time, interfacial processes in metallic systems during freezing or even during solid-solid transformations. The x-ray transmission microscope (XTM) operates in the hard x-ray range (10 to 100 keV) and achieves magnification through projection. We have obtained, using select aluminum alloys, in-situ records of the evolution of interface morphologies with characteristic lengths as small as 25 μm, interfacial solute accumulation (fig.129) and formation of droplets.

With metallic and semiconducting samples, the penetration of macroscopic layers requires photon energies in excess of 10 keV. This precludes the use of optical approaches for imaging. Only projection radiography can be practically employed in this energy range of over 10 keV. Projection radiography uses the divergence of the beam from a small source. The ultimate resolution is limited by the diameter of the source.

The major components of the system include a metal sample (thickness of order

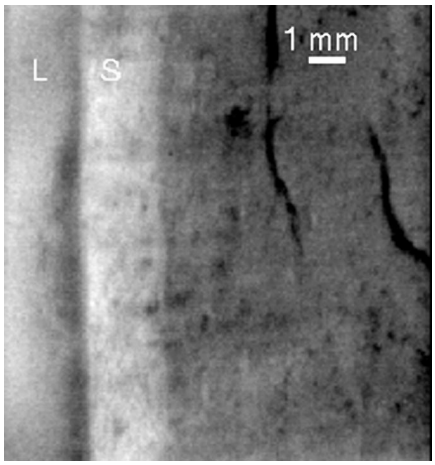

FIGURE 129.—Al-18 in solid/liquid interface growing at 12.4 μm/sec in a 45 °C/cm temperature gradient showing solute rejection to the liquid after the step increase of translation rate.

mm) which is contained in a specially designed, high-transmittance crucible and a high-temperature furnace on a translation stage imposes a temperature gradient onto the sample. The solid-liquid interface is positioned in close proximity to the focal spot of a microfocus x-ray source. The diverging x-ray beam permeates the sample and the resulting shadow falls on an x-ray image converter. The resulting visible image is converted to a digital image by a CCD camera and stored in a computer. This image is displayed on a high resolution monitor, either in real-time or after further processing (contrast enhancement, filtering, etc.).

At typical solidification rates, motion-induced blurring limits the exposure time to a few seconds. With state-of-the-art x-ray image intensifier/camera combinations, a magnification on the detector of some 20 times is the minimum required to obtain a spatial resolution of 10 μm. Such resolution is needed to see the dendritic structures formed in solidifying metals.

Of course such observations require sufficient contrast (difference in absorptance) between features to be resolved and the retention of this contrast by the imaging devices (x-ray converter, image intensifier, camera, recording device). We found that employing higher magnifications than required by simple resolution arguments, provide an improved response in detecting low-contrast features. In monocomponent metallic systems, contrast between solid and melt is determined by the (electron cloud) density of the two phases resulting in less than 2 percent radiographic (image) contrast. In alloy systems, solute segregation can provide further contrast enhancement. The magnitude of contrast is proportional to the difference in atomic number of the components and their concentration. We therefore select the alloys based on x-ray contrast, and we employ the highest magnifications practicable.

How much of the original image contrast is retained depends on the dynamic range of the detector (imaging train) and the size of the features in the object. For small length scales, the contrast retained by the imaging train becomes much smaller than the original image contrast. This can only be (partly) compensated for if the dynamic range of the imaging train is high enough and if the lowest intensities of interest remain above the noise of the system.

We have been using a conventional x-ray image intensifier coupled to cooled (visible light) CCD camera of 12 to 16 bits dynamic range. The intensifier offers at best, a 10-bit dynamic range (1 part in 1,000). Evaluation of new CCD x-ray converter and camera technology was performed using radiation hardened CCD's as a direct conversion, hard x-ray detector. Comparisons between these and phosphor-coated CCD's were used to determine the best technologies to view the low-contrast details of solidification.

Research goals include studying solidification of metals and semiconductors and the dispersion of reinforcement particles in

FIGURE 130.—Optical intensity (absorption) profile along the line in A) crossing the solute layer and interface. Solute gradient is clearly seen on the left part of the graph. Increasing in content represents increased absorption. The diffuse interface region is in the marked area. Note the solute layer is not uniform along the length of the interface.

composites. Features we have already observed include dendrites and cells, the effects of voids and particles on the morphology during solidification of metal matrix composites, and solutal segregation profiles.

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